

Optical Anisotropy of Electronic Excitations in Elliptical Quantum Dots

Achintya Singha,^{1,*} Vittorio Pellegrini,¹ Sokratis Kalliakos,² Biswajit Karmakar,¹ Aron Pinczuk,^{3,4} Loren N. Pfeiffer,⁴ and Ken W. West⁴

¹*NEST INFM-CNR and Scuola Normale Superiore, Pisa 56126, Italy*

²*Department of Materials Science and Technology,
University of Crete, P.O. Box 2208, 71003 Heraklion Crete, Greece*

³*Depts of Appl. Phys & Appl. Math. and of Physics,
Columbia University, New York 10027, USA*

⁴*Bell Laboratories, Alcatel-Lucent, Murray Hill, New Jersey 07974, USA*

(Dated: October 31, 2008)

Abstract

The authors report that anisotropic confining potentials in laterally-coupled semiconductor quantum dots (QDs) have large impacts in optical transitions and energies of inter-shell collective electronic excitations. The observed anisotropies are revealed by inelastic light scattering as a function of the in-plane direction of light polarization and can be finely controlled by modifying the geometrical shape of the QDs. These experiments show that the tuning of the QD confinement potential offers a powerful method to manipulate electronic states and far-infrared inter-shell optical transitions in quantum dots.

PACS numbers: 68.65.Hb, 78.67.Hc, 71.70.-d

Electronic states in circular quantum dots (QDs) are characterized by rotational symmetry that gives rise to an atomic-like shell structure in which states with opposite angular momentum are degenerate [1]. A convenient description of single-particle QD levels is provided by the Fock-Darwin (FD) orbitals with energies given by $E_{nm} = \hbar\omega_o(2n + m + 1) = \hbar\omega_o(N + 1)$, where $n=0,1,\dots$, $m=0,1,\dots$ are the radial and azimuthal quantum numbers, respectively, and ω_o is the in-plane confinement energy. The studies of the spin and charge configurations in QDs by transport and optical techniques have been a major research theme in modern condensed matter with several implications in the field of quantum computation [2, 3, 4]. QDs in the low-density limit are also fascinating nanostructures. In this limit, the breakdown of the FD scheme leads to correlated configurations that reveal the impact of fundamental electron interaction [5].

The shell structure of circular QDs manifests in the spectra of neutral collective modes probed by inelastic light scattering [6]. The light scattering experiments are able to probe both spin and charge inter-shell monopole excitations with even change of ΔN in the FD shell and without change in angular momentum, as required by light scattering selection rules. In case of charge inter-shell excitations, the selection rules dictates that the polarizations of the incoming and scattered photons have to be parallel. In circular QDs, these electronic charge excitations are isotropic in the plane i.e. their energy does not depend on the direction of the polarizations of the incoming and scattered photons, which reflects the rotational symmetry of the nanostructure.

In this letter we demonstrate that asymmetric QDs with few electrons present a peculiar optical anisotropy that manifests in their inter-shell excitation spectra. By nanofabrication we designed different AlGaAs/GaAs quantum dot structures composed by two closely-spaced QDs having the overall lateral shape like the one shown in Fig.1. The two main parameters characterizing our nanostructures are the diameter $2R$ of the two QDs and the inter-dot distance D . The lateral anisotropy is gradually modified by increasing the value of D .

We measured the low lying collective inter-shell charge excitations with polarizations along and perpendicular to the inter-dot axis by resonant inelastic light scattering. The spectra reveals a large energy-splitting between the excitations in the two configurations. The evolution of energy-splitting in the two polarization configurations in samples with different D values represent a direct proof of the breakdown of rotation invariance and consequent lifting of the degeneracies of the single-particle levels present in a circularly-shaped

dot. In addition, the observed anisotropy in the scattered light implies the breakdown of the light scattering selection rules that could be induced by heavy-light hole mixing due to the asymmetric confinement. We remark that the impact of ellipsoidal deformation on the Coulomb blockade transport properties has been studied as a function of a magnetic field [7] but, to the best of our knowledge, no direct experiments probing the optical anisotropy of electronic excitations has been reported so far. This is particularly striking since ground states, selection rules and matrix elements of both dipole and multipole transitions of elliptically-shaped QDs have been extensively calculated [8, 9, 10, 11, 12, 13, 14] as well as valence band mixing effects and interband optical properties [15]. In the single electron picture, the conduction-band energy level structure of the elliptical QD is given by $E_{n_X n_Y} = \hbar\omega_X(n_X + 1/2) + \hbar\omega_Y(n_Y + 1/2)$ [11] where ω_X and ω_Y represents the confinement energies in the X and Y directions respectively, as shown in Fig.1 (right-upper panel). Studies of photoluminescence polarization anisotropies have been also reported particularly in nanorod crystals [16].

Samples were fabricated from a 25 nm wide, one-side modulation-doped $\text{Al}_{0.1}\text{Ga}_{0.9}\text{As}/\text{GaAs}$ quantum well with measured low-temperature electron density $n_e = 1.1 \times 10^{11} \text{ cm}^{-2}$ and a mobility of $2.7 \times 10^6 \text{ cm}^2/\text{Vs}$. QDs were produced by inductive coupled plasma reactive ion etching. QD arrays (with sizes $100 \times 100 \mu\text{m}$ containing 10^4 single identical QD replica) were defined by electron beam lithography. Deep etching (below the doping layer) was then achieved. The lateral asymmetry is introduced by fabrication of two identical quantum dots, with identical radii R , close to each other having centre to centre distance D . When the distance D is less than $2R$ the confinement resembles that of a single quantum dot with an elliptical-like potential (see Fig.1). As D increases towards values $D > 2R$, the two quantum dots tend to decouple into two isolated circular QDs (see Fig.3). Here we focus on QDs with $R = 90 \text{ nm}$ and values of D in the range between 170 nm and 260 nm. Thanks to the presence of a depletion layer, the effective confinement (green shaded region in Fig.1) is less than the geometrical values. In the uncoupled regime at large D the estimated population of each QDs is 3-5 electrons and $\omega_Y \approx 4 \text{ meV}$ [17].

The experiments were performed in a backscattering configuration at temperatures $T = 1.9 \text{ K}$. A tuneable ring-etalon Ti:Sapphire laser was focused on a $100 \mu\text{m}$ -diameter area for excitation of the quantum dots array and the scattered light from the quantum dots array having the same polarization direction of the incoming light was collected through a series

of optics, dispersed by a triple-grating spectrometer and detected by a CCD multi-channel detector. A polarization rotator between the laser and the sample and a $\lambda/2$ plate in front of the spectrometer were used to define the two polarization configurations: XX-both incident and scattered light are parallel to the inter dot axis and YY-the same for perpendicular direction.

The right-bottom panel in Fig.1 shows representative inelastic light scattering spectra of inter-shell charge excitations in the two polarization configurations. The spectra reveal two distinct excitations (A and B) that are selectively observed in the two polarization configurations YY and XX, respectively. The thick blue and red lines are Gaussian fits to the experimental data. The observed energy difference between the peak A and B is the signature of the large optical anisotropy due to the asymmetry of the in-plane confinement potential.

The manifestation of the optical anisotropy in light scattering is intriguing. In a cylindrical QD the electronic states can be labelled by their angular momentum quantum numbers. Therefore interband transitions involving both light-hole and heavy-hole states should be isotropic in the X-Y plane i.e. do not depend on the polarization direction in X-Y plane. The deviation from this behavior in our QDs could be explained by invoking mixing between heavy- and light-hole states induced by the external asymmetric potential. While a quantitative calculation of this effect is beyond the scope of this paper, we note that the origin of optical anisotropies in asymmetric nanostructures and the role of valence-band mixing have been theoretically addressed in several works [15, 18, 19, 20].

The energy splitting of the modes A and B that is around 2 meV, can be related to $\omega_Y - \omega_X$ although their absolute energies are expected to deviate from ω_Y and ω_X due to the impact of electron correlations that we expect significant in these regimes of low electron occupation [17]. The splitting between the inelastic light scattering resonance enhancement peaks in the XX and YY configurations shown in Fig.2 is also close to 2 meV. This fact suggests small splittings of valence band states of the X and Y inter-band transitions involved in the light scattering process. If we consider $\omega_Y - \omega_X \approx 2$ meV, and $\omega_Y \approx 4$ meV [17] we deduce $\omega_X \approx 2$ meV and therefore an ellipticity factor $\delta = \omega_X/\omega_Y = 0.5$ for the QD shown in the inset to Fig.1.

To demonstrate tuning of the optical anisotropy we report in Fig.3 the evolution of the charge excitation spectra in both XX and YY configurations as a function of the inter-dot

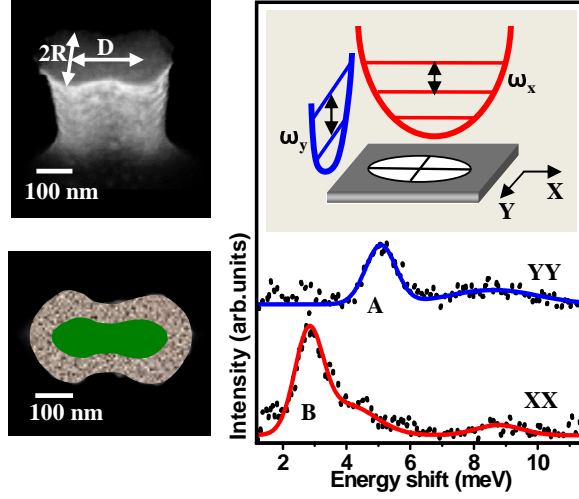


FIG. 1: SEM images of coupled quantum dot. The semi-elliptical green shaded area represents the effective confinement region of the electrons. This coupled system clearly shows the anisotropy of the confinement potential. XX represents the configuration of charge excitation when both incident and scattered light are parallel to the inter dot axis. YY represents the same when they are perpendicular to the inter dot axis. The energy parabola along the major and minor axes are included and the respective excitations A and B, are marked. Inelastic light scattering charge excitation spectra for the light polarization configuration XX and YY. The spectra are shifted vertically for clarity.

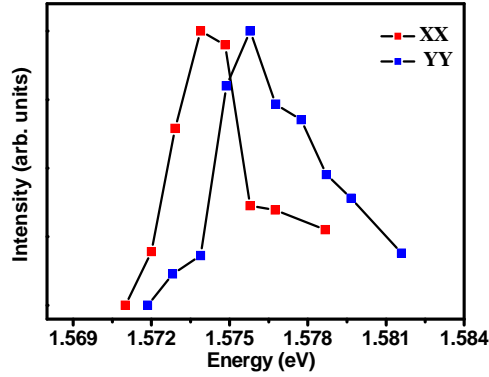


FIG. 2: Inelastic light scattering resonant enhancement profiles of the inter-shell charge collective modes in the two polarization configurations.

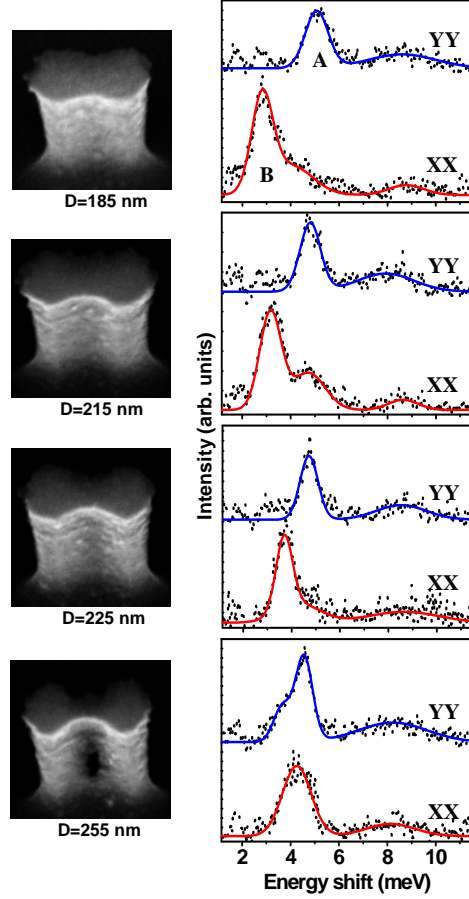


FIG. 3: Left vertical panels: SEM images of the coupled QDs with different inter-dot distance D . Right vertical panels: charge excitation spectra for XX and YY polarization configuration.

distance D . The results displayed in Fig.3 indicate that the energy difference between peaks A and B decreases with the increase of D and, as expected, it vanishes in the case of nearly decoupled dots when $D > 2R$. In this case the re-established circular symmetry leads to similar charge spectra in the two polarizations. Finally Fig.4 reports the A-B energy splitting values as a function of inter-dot distance. The red line is an exponential fit to the data with the function $A(e^{-D} - e^{-D_0})$ where $D_0 \approx 270$ nm.

In conclusion, we have shown that the tuning of the QD confinement potential offer a method to manipulate electronic states with great precision. The breakdown of rotational invariance here reported might offer new venues for the exploration of electron correlation effects in these nanostructures. In addition, the anisotropic QDs here studied might be usable for polarization-dependent detection of far-infrared radiation.

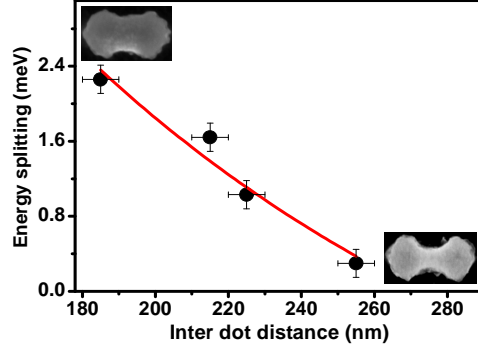


FIG. 4: Evolution of the energy splitting between the two chargemodes (A-B) in YY and XX polarization configurations as a function of the inter-dot distance. The red line is an exponential fit to the data. The insets show SEM images of the QDs corresponding to the two extreme points.

This work was supported by the projects MIUR-FIRB No. RBIN04EY74, A. P. is supported by the National Science Foundation under Grants No. DMR-0352738, DMR-0803691 and CHE-0641523. By the Department of Energy under Grant No. DE-AIO2-04ER46133, and by a research grant from the W. M. Keck Foundation. V.P acknowledge partial support of the Italian Academy and NSEC at Columbia University. We acknowledge useful discussion with Massimo Rontani, Guidi Goldoni and Elisa Molinari.

* Electronic address: a.singha@sns.it

- [1] S. M. Reimann and M. Manninen, Rev. Mod. Phys. **74**, 1283 (2002).
- [2] L. P. Kouwenhoven, T. H. Oosterkamp, M. W. S. Danoesastro, M. Eto, D. G. Austing, T. Honda, and S. Tarucha, Science **278**, 1788 (1997).
- [3] R. Hanson, L. P. Kouwenhoven, J. R. Petta, S. Tarucha, and L. M. K. Vandersypen, Rev. Mod. Phys. **79**, 1217 (2007).
- [4] X. Xu, B. Sun, P. R. Berman, D. G. Steel, A. S. Bracker, D. Gammon, and L. J. Sham, Science **317**, 929 (2007).
- [5] S. Kalliakos, M. Rontani, V. Pellegrini, C. P. Garcia, A. Pinczuk, G. Goldoni, E. Molinari, L. N. Pfeiffer, and K. W. West, Nature Phys. **4**, 467 (2008).
- [6] C. Schuller, K. Keller, G. Biese, E. Ulrichs, L. Rolf, C. Steinebach, D. Heitmann, and K. Eberl

- Phys. Rev. Lett. **80**, 2673 (1998).
- [7] Y. Tokura, S. Sasaki, D. G. Austing, and S. Tarucha, Physica B **298**, 260 (2001).
 - [8] M. Marlo, A. Harju, and R.M. Nieminen, Phys. Rev. Lett. **91**, 187401 (2003).
 - [9] G. Cantele, D. Ninno, and G. Iadonisi, Nano Lett. **1**, 121 (2001).
 - [10] L. Serra, A. Puente, and E. Lipparini, International J. of Quantum Chemistry **91**, 483 (2003).
 - [11] A. V. Madhav and T. Chakraborty, Phys. Rev. B **49**, 8163 (1994).
 - [12] L-L. Sun, F-C Ma, and S-S Li, J. Appl. Phys. **94**, 5844 (2003).
 - [13] D. Xu and J-L Zhu, Phys. Rev. B **72**, 075326 (2005).
 - [14] Y. Li, C. Yannouleas, and U. Landman, Phys. Rev. B **76**, 245310 (2007).
 - [15] S. Cortez, O. Krebs, P. Voisin, and J. M. Gerard, Phys. Rev. B **63**, 233306 (2001).
 - [16] J. Hu, L-s Li, W. Yang, L. Manna, L-w Wang, A. P. Alivisatos, Science **292**, 2060 (2001).
 - [17] C. P. Garcia, V. Pellegrini, A. Pinczuk, M. Rontani, G. Goldoni, E. Molinari, B. S. Dennis, L. N. Pfeiffer, and K. W. West, Phys. Rev. Lett. **95**, 266806 (2005).
 - [18] L. Besombes, K. Kheng, and D. Martrou, Phys. Rev. Lett. **85**, 425 (2000).
 - [19] O. Krebs and P. Voisin, Phys. Rev. Lett. **77**, 1829 (1996).
 - [20] W. Shenga, Appl. Phys. Lett. **89**, 173129 (2006).